AFRL-AFOSR-JP-TR-2017-0010

Nano-Scale Devices for Frequency-Based Magnetic Biosensing

Peter Metaxas
UNIVERSITY OF WESTERN AUSTRALIA

01/31/2017
Final Report

Air Force Research Laboratory
AF Office Of Scientific Research (AFOSR)/ IOA
Arlington, Virginia 22203
Air Force Materiel Command

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YYYY)*	2. REPORT TYPE	3. DATES COVERED *(From - To)*
09-02-2017	Final	05 Jun 2015 to 04 Nov 2016

4. TITLE AND SUBTITLE
Nano-Scale Devices for Frequency-Based Magnetic Biosensing

5a. CONTRACT NUMBER

5b. GRANT NUMBER
FA2386-15-1-4058

5c. PROGRAM ELEMENT NUMBER
61102F

6. AUTHOR(S)
Peter Metaxas

5d. PROJECT NUMBER

5e. TASK NUMBER

5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
UNIVERSITY OF WESTERN AUSTRALIA
35 STIRLING HIGHWAY
CRAWLEY, 6009 AU

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AOARD
UNIT 45002
APO AP 96338-5002

10. SPONSOR/MONITOR'S ACRONYM(S)
AFRL/AFOSR IOA

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
AFRL-AFOSR-JP-TR-2017-0010

12. DISTRIBUTION/AVAILABILITY STATEMENT
A DISTRIBUTION UNLIMITED: PB Public Release

13. SUPPLEMENTARY NOTES

14. ABSTRACT
We demonstrate via experiment and simulation that the magnetic-field-dependent frequency of resonant magnetization dynamics in ferromagnetic nanostructures can be exploited for frequency-based detection of magnetic fields generated by magnetic nanoparticles and microbeads. The electronic detection of these fields is of high practical interest since suitably functionalized magnetic particles can be used used as detection-enabling labels for biological analytes (e.g. disease markers), opening pathways toward the development of solid state biological sensors (biosensors). We have demonstrated magnetic particle detection using ferromagnetic resonances in both large magnonic crystals and, via electrical measurements, in magnetic-vortex-containing, isolated micro- and nano-devices. Via micromagnetic simulations, we have largely determined the mechanisms underlying the observed nanoparticle-driven shifts. This resonance-based approach is particularly promising for the development of fast, frequency-based electronic sensors for nanoparticles and microbeads which can operate in relatively high magnetic fields (100s of mT) and can be scaled down to have lateral dimensions on the order of 100 nm and below.

15. SUBJECT TERMS
biosensing, magnetic nanoparticles, protein markers

16. SECURITY CLASSIFICATION OF: a. REPORT	b. ABSTRACT	c. THIS PAGE	17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
Unclassified	Unclassified	Unclassified	SAR	18	AHLERS, KRISTOPHER
					19b. TELEPHONE NUMBER *(Include area code)* 315-227-7009

"NANO-SCALE DEVICES FOR FREQUENCY-BASED MAGNETIC BIOSENSING"
Final Report for AOARD Grant FA2386-15-1-4058

4th February 2017

PI information: Peter Metaxas; peter.metaxas@uwa.edu.au; University of Western Australia; School of Physics; M013, 35 Stirling Highway, Crawley WA 6009; +61 8 6488 7015; Fax.

Period of Performance: 06/05/2015 – 11/04/2016

Abstract: We demonstrate via experiment and simulation that the magnetic-field-dependent frequency of resonant magnetization dynamics in ferromagnetic nanostructures can be exploited for frequency-based detection of magnetic fields generated by magnetic nanoparticles and microbeads. The electronic detection of these fields is of high practical interest since suitably functionalized magnetic particles can be used used as detection-enabling labels for biological analytes (e.g. disease markers), opening pathways toward the development of solid state biological sensors ("biosensors"). We have demonstrated magnetic particle detection using ferromagnetic resonances in both large magnonic crystals and, via electrical measurements, in magnetic-vortex-containing, isolated micro- and nano-devices. Via micromagnetic simulations, we have largely determined the mechanisms underlying the observed nanoparticle-driven shifts. This resonance-based approach is particularly promising for the development of fast, frequency-based electronic sensors for nanoparticles and microbeads which can operate in relatively high magnetic fields (100's of mT) and can be scaled down to have lateral dimensions on the order of 100 nm and below.

Introduction: NB: Below, references appearing in square brackets "[]" refer to papers resulting from this project which are listed toward the end of the main part of the report.

Magnetic biosensing methods use magnetic nanoparticle or microbeads as detection-enabling labels for biological analytes[1]. The absence of a magnetic background in most biological samples makes this sensing approach highly versatile, opening promising pathways towards portable, robust tools for medical diagnostics or environmental monitoring which are based on solid state sensors. Quite a large focus has historically been given to giant or tunneling magnetoresistive sensors for nanoparticle detection[1]. Sensing with such devices is based on detecting particle-induced changes to the (quasi-)static magnetization within the active layer of the device[2]. This project however focuses on particle-induced changes to high frequency magnetization dynamics. See (e.g.) Fig. 1 in [3].

The ultimate goal of this research has been to identify, model, understand and optimize nanoparticle-induced modifications to high frequency magnetization dynamics in magnetic nanostructures with a view to develop the science and technology behind next generation nanomagnetic biosensing devices. By exploiting magnetization *dynamics*, it has been predicted that it will be possible to shrink sensor sizes to around 100 nm while transitioning to a detection modality which is intrinsically frequency-based (rather than voltage-level based, as is the case

[1] H. Lee et al., Chemical Reviews, 115, 10690 (2015).
[2] J.-R. Lee et al., Scientific Reports, 6, 18692 (2016).

for conventional magnetoresistive sensors)[3]. This sensing approach can also be used in the presence of relatively large magnetic fields (system dependent but on the order of 0.1 – 1 T). The latter enables an increase in the particle moment and thus the generated stray fields which are the basis of particle detection.

Specifically, the aims of the project (as stated in the agreement) were as follows:
1. Probe nanoparticle-induced changes to nano-confined modes of resonant magnetization dynamics in an easily-probed, large area system under various external magnetic field strengths and orientations.
2. Probe nanoparticle induced changes to the resonance of the magnetic vortex state in a simple, isolated micro-structure.
3. Demonstrate frequency-based sensing with a direct-current-driven nanodevice.

This project has involved collaborators at the University of Western Australia (including MPhys and PhD students as well as a listed key researcher, Mikhail Kostylev), the University of Southampton (UK; key researcher: Prof Hans Fangohr), AIST (Japan), the Unité Mixte de Physique CNRS/Thales (France; key researcher: Dr Vincent Cros) and the National University of Singapore (Singapore; key researcher Prof Adekunle Adeyeye).

Experiment: The main goal of the experiments and simulations was to characterize and interpret particle-induced FMR frequency shifts in magnetic nanostructures. In large area magnonic crystals, resonances were identified experimentally using broadband ferromagnetic resonance spectroscopy (see [3] and references therein). The gyrotropic resonance in vortex-containing disks was studied experimentally via magnetoresistive rectification measurements (see [4]). In STOs two methods were used which are detailed below. Note that STO work greatly benefited from a new measurement setup (partly funded by the AOARD and constructed during the performance period) which allowed high throughput device testing using radiofrequency probes and a projected field electromagnet (Fig. 1).

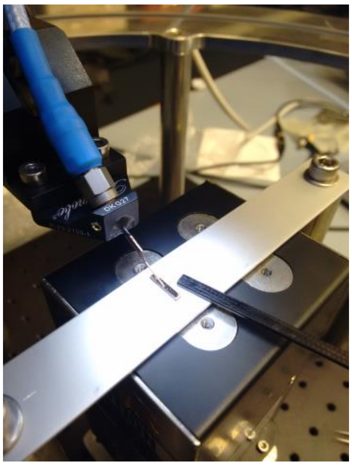

Fig. 1. Close-up image of the new magnetotransport setup fabricated and commissioned during this project. A radiofrequency probe is used to electrically contact a spin torque oscillator on a small rectangular silicon wafer which is mounted above one of the (silver) poles of a projected field electromagnet. The black probe is a Hall probe for measuring the field.

[3] P.M. Braganca et al., Nanotechnology, 21, 235202 (2010).

Resonances were simulated either via a magnetic eigenmode approach using the FinMag code (developed by Prof Hans Fangohr's team) or via time-resolved micromagnetic simulations run using the open source, GPU-based Mumax3 code (GPU = graphics processing unit). More details on the simulation methods (including techniques to model the nanoparticles) have been outlined in publications [1-7].

Particle positions were determined experimentally via scanning electron microscopy (SEM).

Results and Discussion: Below, results are separated by the above listed aims. Potential future work is summarized at the end of this section.

Aim 1

The first aim of the project was focused on magnonic crystals consisting of nanoscale holes within a magnetic film: "probe nanoparticle-induced changes to nano-confined modes of resonant magnetization dynamics in an easily-probed, large area system under various external magnetic field strengths and orientations." Over the course of this project, we completed a highly detailed study of resonance-based detection in finite magnetic fields (1a) and carried out frequency-resolved measurements which enabled particle sensing in the absence of a static external magnetic field (1b). Studies of particle sensing in the presence of out-of-plane fields (1c) are continuing.

1a. Demonstrating particle detection for a wide range of particle sizes using nanoconfined ferromagnetic resonances in a large area magnetic nanostructure

Associated paper: [3] M. Sushruth, J. Ding, J. Duczynski, R.C. Woodward, R.A. Begley, H. Fangohr, R.O. Fuller, A.O. Adeyeye, M. Kostylev, P.J. Metaxas, "Resonance-based Detection of Magnetic Nanoparticles and Microbeads Using Nanopatterned Ferromagnets", Physical Review Applied, 6, 044005 (2016).

In [3], via experiment and micromagnetic simulation, we demonstrated that nanoconfined ferromagnetic resonances can be used to detect a wide range of particle sizes, ranging from 6 nm to 4 microns (i.e. over ~ 3 orders of magnitude). We also explicitly show that nanoparticle detection can be successfully carried out at quite high magnetic fields where magnetic particle moments and, as a result, the measured resonance shifts are maximized (see Fig. 5 in [3]). The maximum tested field was ~0.3 T (see supplementary information for [3]).

A large emphasis was given to particles with widths on the order of 130 nm since these have been well characterized by the PI's team. They also exhibit quite a repeatable particle positioning within the magnonic crystal's array of holes, making them conducive to simulation. For these particles we were able to reproduce, via micromagnetic simulation, the shifts seen at various magnetic fields for two particle coverages which were amenable to simulation: a coverage corresponding to ~1 particle per hole and a coverage where the entire nanostructure was covered by a semi continuous layer of particles (Fig. 4c in [3]). We also explicitly show in this work (again via simulation *and* experiment; Figs. 2 and 4c in [3]) that the shifts are significantly lower if sensing is carried out using a continuous (i.e. unpatterned) film. We note that this is not due to an intrinsically low sensitivity of the resonance frequency to field but rather due to the fact that in the patterned system particles can be captured in the holes and thus are consistently positioned close to the modes which are confined in the nanoscale regions surrounding each holes. Finally,

we also discuss the source of the differing shift polarities induced by small particles which can enter the magnonic crystal's holes versus those induced by large particles which sit on top of the nanopatterened structure (Sec. IIIC in [3]).

1b. Using magnonic crystals for particle sensing in the absence of an external, static magnetic field

The particles that are being sensed are typically superparamagnetic. Due to thermal fluctuations, their magnetic orientation fluctuates in time and they exhibit a finite time-averaged moment only if there is an external field applied. Although applying large fields during sensing can be advantageous (i.e. by increasing the particle stray field and thus the frequency shifts, as discussed above), field generation can complicate device design due to the need to include a permanent magnet, electromagnet or coil into the device.

Li et al.[4] have however shown that it is possible to sense magnetic particles (in their case using a more conventional giant magnetoresistive sensor) in the absence of an applied field. They did this by creating a groove-shaped void in the sensor. There, the discontinuity in the ferromagnetic sensing element generates a stray field which magnetizes the nanoparticles, thereby leading to a finite particle-generated field which enables particle detection.

We have run experiments to examine the potential for zero-field sensing with the magnonic crystals (which contain also contain voids: the nano-scale holes). Although our measurements typically are field resolved and run at fixed frequency [3] (albeit enabling a determination of the associated shift in the resonance frequency shift), here we had to carry out frequency-resolved measurements involving frequency sweeps at a fixed (or null) external field. This enabled us to probe the resonance *without* changing the field, thus enabling us to access the resonances at zero field.

Encouragingly, we consistently observe particle-generated frequency-shifts at zero external field and note that this was confirmed with particles from two different suppliers. However, the shifts are larger than expected if we assume that it is only the field within the magnonic crystal's holes (generated by the magnonic crystal itself) which is magnetizing the particles. A complete analysis of existing results (including micromagnetic simulations) as well as the collection of additional data sets (to ensure reproducibility) is continuing in 2017.

In Fig. 2 we do however show ferromagnetic resonance traces where the frequency of the "extended mode" (see [3] for more details on this mode) is experimentally observed to shift upwards (on average) for increasing particle coverages in the absence of a static, externally applied magnetic field (particles are PrecisionMRX particles from Senior Scientific). Scanning electron microscopy images in the same figure show the coverages following applications of the lowest ("conc1") and 3rd highest ("conc3") particle solutions. At "conc1" we observe clear chaining effects which will need to be considered in the final analysis since they may stabilize the particles' moments to some degree in the absence of a large external field. We must also consider potential effects from the weak modulation field used in the experiment as well as any other potential parasitic fields at the sample location. We also plan to carry out experiments on continuous (unpatterned) layers. If similar shifts are observed there, that will suggest that the

[4] Y. Li et al., Applied Physics Letters, 104, 122401 (2014).

field within the holes of the nanopatterned magnonic crystal plays little role in magnetizing the particles (i.e. the shifts may instead be related to chaining or other effects).

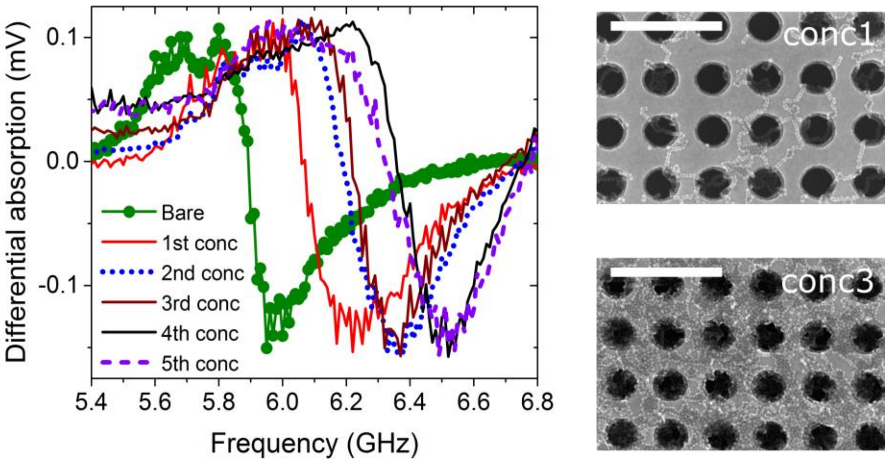

Fig. 2. Ferromagnetic resonance spectra obtained for the "extended mode" (see [3] for details) at zero external field in a magnonic crystal with 300 nm wide holes and an array pitch of 450 nm. Traces are for the bare magnonic crystal and following the application (in zero external field) of increasingly concentrated solutions of magnetic nanoparticles (measurements were carried out after the solution was dried). Images on the right are SEM images taken after the application of the 1st and 3rd solution concentrations (conc1 and conc3). The scale bar is 1 micrometer long.

1c. Sensing in the presence of out-of-plane fields

In [3], discussed in section 1a, nanoparticle sensing was carried out with the field applied in the plane of the film. One of the proposed activities in this project was to also carry out equivalent experiments in the presence of an out-of-plane field where, for sufficiently high fields, the resonance frequency is expected to have a linear dependence on field. This work commenced at the end of 2016 when a dedicated Honours student was recruited specifically for this part of the project. The research is now ongoing. Some simulation results from early in the project are however shown in Fig. 3. In contrast to the case of an in-plane field [3], all of the simulated modes shift downward in frequency in the presence of a nanoparticle (red line). This result is consistent with the out-of-plane component of the magnetic field generated by a hole-located particle being oriented *against* the applied field within the magnetic part of the magnonic crystal. This would act to reduce the total field acting on the magnetic moments within the magnonic crystal and thus reduce the resonance frequency.

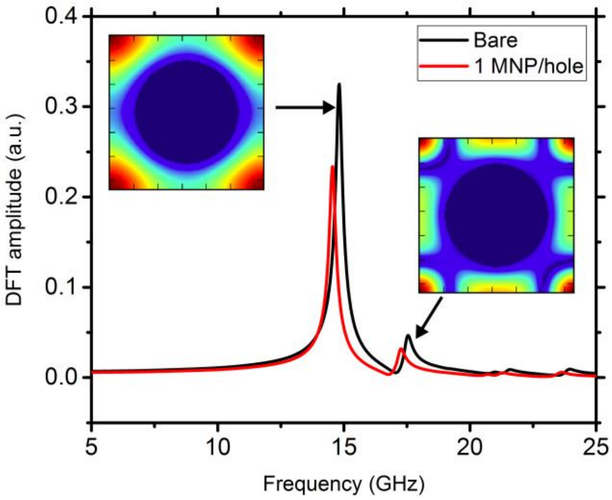

Fig. 3. Simulated resonance spectra for a magnonic crystal with 300 nm wide holes and a 450 nm array pitch (both bare and with a 150 nm wide iron oxide nanoparticle in each hole). Insets show the concentration of the dynamics for the two lowest frequency modes (red = high amplitude dynamics).

Aim 2

The second aim of the project was to "probe nanoparticle induced changes to the resonance of the magnetic vortex state in a simple, isolated micro-structure." This part of the work grew in significance over the course of the project thanks to some new findings coming from both experiments and simulations on vortices in isolated disks. These included (2a) the first detailed, micromagnetism-based explanation of the nonlinear field dependence of the vortex gyrotropic resonance frequency in small nanostructures; (2b) the prediction of a confinement-enhanced field sensitivity (relevant to spatially localized magnetic fields such as those generated by nanoparticles); and, (2e,f) novel routes for detecting vortex core polarity switching and for enhancing gyrotropic frequency tuning. These results are in addition to those focused on experimental particle detection (2d) and simulation-based particle detection optimization (2c).

2a. Nonlinear field-dependence of the gyrotropic resonance frequency

Associated paper: [1] J.P. Fried, H. Fangohr, M. Kostylev, P.J. Metaxas, "Exchange-mediated, non-linear, out-of-plane magnetic field dependence of the ferromagnetic vortex gyrotropic mode frequency driven by core deformation", Physical Review B, 94, 224407 (2016).

The gyrotropic resonance frequency is typically considered to have a linear dependence on the magnitude of an external, out-of-plane magnetic field[5]. This dependence thus has the potential to be directly exploited for detecting fields that are spatially quite uniform across the sensor. Examples of such fields include those generated by microbeads which have can have widths which are an order of magnitude wider than the vortex-containing device.

However, results from two studies published in 2010 and 2016 showed that the gyrotropic

[5] G. de Loubens et al., Physical Review Letters, 102, 177602 (2009).

frequency could actually decrease when approaching the disk's saturation field (this is the field at which the vortex state is destroyed and the disk becomes uniformly out-of-plane magnetized). This non-linearity can complicate field sensing since the frequency no longer shows a monotonic dependence on field. Furthermore, this non-linearity is strongest in devices with small lateral dimensions and is thus relevant to nano-scale sensor development, one of the motivations for this work.

Via analysis of micromagnetic simulation results we were able to carry out a detailed analysis of the nonlinearity (Fig. 1(a) in [1]). We showed that it is due to a deformation of the shifted vortex's structure which is driven by non-uniform out-of-plane demagnetizing fields (Fig. 2av in [1]). The deformation is such that it reduces the exchange energy cost of the core displacement, effectively reducing the exchange-mediated component of the core stiffness and thus the core's gyrotropic frequency. The nonlinearity is highest in small devices since the relevant part of the demagnetizing field profile becomes more non-uniform as the disk width is decreased, generating a higher degree of deformation. This paper is also one of the first to highlight the important role played by core deformation in determining exchange contributions to the gyrotropic frequency.

2b. Confinement-driven frequency-shifting of the gyrotropic resonance frequency

Associated paper: [5] J.P. Fried and P.J. Metaxas, "Localized magnetic fields enhance the field-sensitivity of the gyrotropic resonance frequency of a magnetic vortex", Physical Review B, 93, 064422 (2016).

As noted above, sufficiently below the disk saturation field, the gyrotropic frequency is a linear function of the out-of-plane field magnitude, enabling the definition of a field sensitivity (i.e. change in frequency per unit field). Via micromagnetic simulations, we have shown that the sensitivity of the gyrotropic frequency to out-of-plane field will be strongly increased if the field is spatially localized and aligned with the magnetization within the vortex core [5]. This increase arises due to the core becoming confined by the localized field. The confinement increases the core stiffness and thus increases the gyrotropic frequency (Fig. 5 in [5]).

An important message to take away from this work is that to predict frequency-shifts generated by spatially *localized* magnetic fields (such as those generated by nanoparticles or other nanostructures), one cannot simply use the field-dependence of the frequency as measured in the laboratory in the presence of uniform fields. Instead we must consider the potential for a confinement-driven contribution to the field-driven frequency shift.

Another consequence of these effects is that when changing the particle size (for a fixed particle-disk) separation, there will be a maximum shift at a critical particle size (Fig. 7 in [5]) where there is an optimal combination of field localization (maximized for small particles) and field magnitude (maximized for large particles). However, below we will discuss some practical limitations to using confinement-driven effects for real world sensing.

2c. Vortex-based particle detection will be most robust for microbeads

Associated paper: [7] J.P. Fried and P.J. Metaxas, "Nanoparticle-modified magnetic vortex dynamics", accepted for publication in IEEE Magnetics Letters (2017) [manuscript # LMAG-16-11-NM-0305.R1].

[7] was recently accepted for publication in IEEE Magnetic Letters and, using micromagnetic

simulation, gives a summary of the different mechanisms underlying vortex-based particle detection. One of the main results is that the confinement-driven frequency-shifting mechanism (detailed above in 2b) becomes weaker for centrally offset nanoparticles, large vortex core orbits and/or significant particle-sensor vertical separations. A conclusion of the paper is that the use of large particles (i.e. microbeads) likely represents a more robust sensing modality if the particles cannot be guaranteed to align centrally over the disk. This is largely due to the fact that microbeads generate stronger, less localized fields than nanoparticles with the resultant dominant frequency-shifting mechanisms (detailed in [7]) being less dependent on particle position and core orbit radius than confinement-driven frequency-shifting.

2d. Particle detection using electrically probed vortex resonance

A major goal of this part of the project was to experimentally demonstrate particle sensing using electrically probed vortex resonances in a single disk. To do this, we exploited magnetoresistive rectification (detailed in [4]) in which radiofrequency currents are driven across a metallic microdisk containing a magnetic layer (which itself contains a magnetic vortex). Magnetoresistive rectification (which depends on anisotropic magnetoresistance) leads to a voltage appearing across the device at the vortex resonance. It thus enables an electrical measurement of the gyrotropic resonance frequency of a single vortex (e.g. Fig. 3(a) in [4]). We have successfully demonstrated particle-induced changes to the gyrotropic resonance frequency in a number of experiments both for magnetic nanoparticles (widths on the order of 0.1 micrometer) and magnetic microbeads (widths of ~ 0.9 – 4 micrometers).

In Fig. 4(a), we show the basic measurement setup (the field is applied perpendicular to the disk plane). A radiofrequency signal is injected across the disk (disks were fabricated by the PI and his collaborators prior to the performance period) with the rectification voltage signal measured using a nanovoltmeter. As in [4], the device resistance versus in-plane magnetic field was confirmed to be characteristic of vortex-containing disks. The rectification peak positions are field-dependent due to the out-of-plane field dependence of the gyrotropic resonance (as discussed in section 2e), enabling us to plot the gyrotropic frequency versus out-of-plane field (Fig. 4b). In Fig. 4b we show that for this particular device, the simulations match the experiment only when including a misalignment of 0.8 degrees between the field and device normal. The inclusion of such an angle (albeit small) proved critical in our reproduction of the experimental results. The angle was likely due to a slightly imperfect alignment of the field and the sample mount or perhaps a slightly angled device wafer.

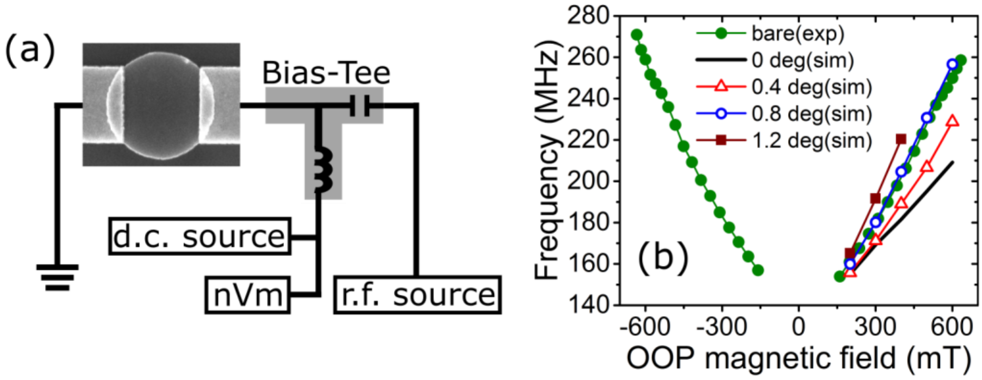

Fig. 4. (a) Top-down SEM image of a 2 micron wide disk (with nonmagnetic contacts) together with a schematic of the measurement circuit (nVm=nanovoltmenter). (b) Experimentally obtained gyrotropic resonance frequencies for the bare disk together with simulated frequencies (positive field only) assuming various values of misalignment (in degrees) between the disk normal and the field.

In Fig. 5 we show results obtained for ~0.9 micron wide polymer-based magnetic beads on a different device (misalignment angle = 1.4 degrees). There are a number of particles near the disk which (Fig. 5a) which consistently generate negative frequency shift (Figs. 5b,c). There is reproducibility of the frequencies via micromagnetic simulations to within a few MHz (Figs. 5c,d). Note that the simulations take the particle positions and the particles' field dependent moments into account. The simulations indicate that the particle stray fields reduce the out-of-plane canting of the magnetization and act to move the vortex core closer to the disk center. Both of these effects are expected to reduce the gyrotropic frequency (see also [7]), consistent with the observations. A paper detailing these results is currently being prepared.

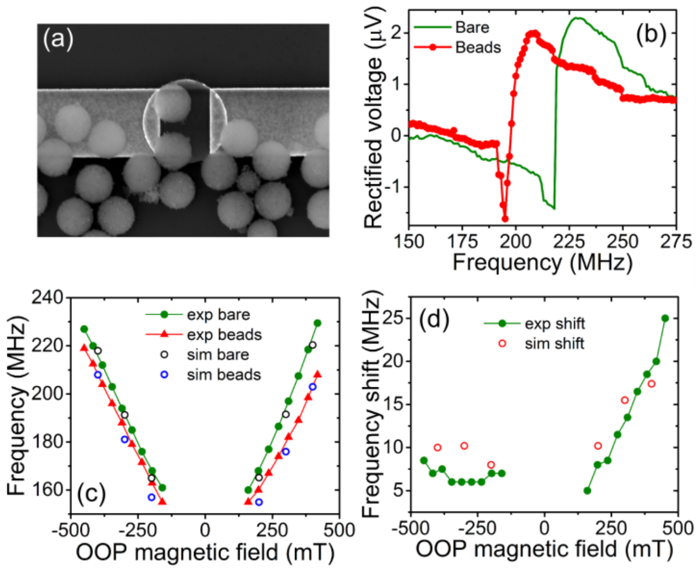

Fig. 5. (a) SEM image of the measured device with magnetic microbeads. (b) Example of the particle-induced shift of the rectification peak. Simulated and experimental field dependence of the gyrotropic frequency with and without particles (c) and the particle-generated frequency shifts (d).

As briefly mentioned above, we have also successfully detected collections of much smaller, isolated nanoparticles on or next to the disks however reproduction of these shifts via simulation is still continuing.

As a final remark, we also note that we successfully ran preliminary experiments wherein we electrically measured vortex gyration while controllably positioning a microbead above a disk using a scanning probe microscope (SPM). This was achieved by attaching a microbead to a SPM cantilever and mounting the modified cantilever within a SPM at UWA (collaboration: Dr Thomas Becker, Curtin University). We note that a dedicated sample holder with electrical connections was fabricated for this experiment. It contains a permanent magnet that allows an approximately out-of-plane field to be applied to the device (and microbead) within the SPM enclosure.

In Fig. 6 we show two magnetoresistive rectification traces obtained inside the SPM (the dip in the voltage trace corresponds to the vortex resonance). We see that a higher frequency is obtained when the tip is directly above the center of the disk than when the tip is retracted and the microbead is far above the disk. This may be consistent with a particle-induced magnetization canting and/or a particle-induced shift of the core towards the edge of the disk. Note that traces have been recorded for various particle locations and they demonstrate the existence of a particle-position-dependent frequency shift (as expected from [7]). The results are thus promising. However, before taking such measurements further we will have to improve the mechanical stability of the setup and take into account the possibility that the field is not perfectly out-of-plane which, as seen above, can strongly affect the resonant frequency and particle-driven shifts. Such work will likely require a dedicated Honours or Masters student in the future. Note that we were not able to identify any detectable vortex-bead interactions in preliminary tests.

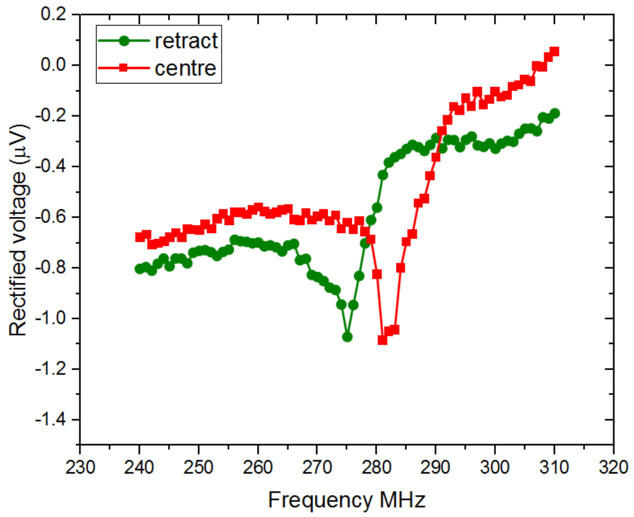

Fig. 6. Magnetoresistive rectification traces obtained while positioning a microbead directly above a disk ("centre") and far above a disk ("retract") using a SPM in the presence of an approximately out-of-plane magnetic field. The gyrotropic resonance is increased in the presence of the bead (identified from the downward dip in the voltage trace).

2e. Magnetoresistive rectification measurements as a probe for vortex core polarity switching

Associated paper: [4] M. Sushruth, J.P. Fried, A. Anane, S. Xavier, C. Deranlot, M. Kostylev, V. Cros, P.J. Metaxas, "Electrical measurement of magnetic-field-impeded polarity switching of a ferromagnetic vortex core", Physical Review B Rapid Communications, 94, 100402(R) (2016).

An unexpected outcome of this work was that we demonstrated that the magnetoresistive rectification measurement method could be employed to identify vortex core polarity switching. Vortex core polarity switching is a highly non-linear dynamic phenomenon of both fundamental and applications-driven interest (see introduction in [4]).

We came across this result when attempting to increase the amplitude of the experimentally measured rectification peak by increasing the driving current. Beyond a certain radiofrequency power however, we found a clear loss of signal within the rectification peak (e.g. Fig. 3c in [4]). Via micromagnetic simulation, we showed that this signal loss was consistent with vortex core polarity switching (e.g. Fig. 1e in [4]). The results were published in Physical Review B Rapid Communications [4] where we also used the technique to demonstrate a route to impede switching. This was based on the field-induced displacement of the core to a steeper part of the core's geometry-induced, anharmonic confining potential.

2f. Enhanced frequency tuning of the gyrotropic resonance.

Associated paper: [8] M. Sushruth, J.P. Fried, A. Anane, S. Xavier, C. Deranlot, V. Cros and P.J. Metaxas, "Chirality-mediated bistability and strong frequency downshifting of the gyrotropic resonance of a magnetic vortex", under review at Physical Review Letters (2017).

Another critical aspect of reproducing the particle shifts (section 2d) is having knowledge of the chirality of the vortex (the direction of the vortex's curling magnetization; see Fig. 1 in [7]). This is important as it defines the direction the core will shift in response to particle generated fields – a shift toward the edge of a circular disk increases the frequency of the gyrotropic mode (e.g. see [7] and references therein).

A fraction of our disks however are not perfectly circular: they have a deliberately flattened edge which enables chirality control and in the later parts of this project we started focusing on these devices given the importance of the chirality to results interpretation. During preliminary measurements, we found that a resonating vortex core which had been shifted toward the flat edge of the disk actually exhibited a decreasing resonance frequency (Fig. 3 in [8]). This is in stark contrast to the frequency increase which occurs when the core is shifted to the round edge of the disk (controlled core shifting can be achieved using in-plane fields). This ability to both increase and decrease the resonance frequency of the core by shifting it to the flat or round edge of the disk enables a strongly enhanced magnetic-field-driven tuning of the gyrotropic mode frequency. We have reproduced the results via simulation, showing that it is due to a reduction in the dynamic core stiffness when the core is moving along the flat edge of the modified disk (Figs. 4a-c in [8]). A paper detailing these results [8] is currently under review at Physical Review Letters.

Aim 3

The final aim of the project was focused on "demonstrat(ing) frequency-based sensing with a direct-current-driven nanodevice" (i.e. a vortex-based spin torque oscillator or STO[6]). In a STO experiment, a direct current is typically driven through the device with an aim to drive magnetization dynamics (in this case vortex oscillations) via spin transfer torque. The STO device is magnetoresistive, leading to oscillations of the device resistance at the frequency of the magnetization dynamics (~ 0.1 – 1 GHz for vortices). This opens new possibilities for real time, high speed particle detection using solid state devices (see reference footnote "3" on page 2).

For aim 3 of this project, we concentrated on multilayer STOs that contained two interacting vortices (see Lebrun et al. for details [7]). Such devices had, at relatively low magnetic fields (20 mT and lower), been shown by Lebrun et al. to be capable of generating signals with low (sub MHz) linewidths which would make them ideal for resolving particle generated frequency shifts.

The measurement setup used in this project for measuring STO outputs is shown in Fig. 7(a). A spectrum analyser (S.A.) (or a high frequency oscilloscope) is used to measure the radiofrequency STO output signal with Fig. 7(b) demonstrating the field dependence of the output signal frequency for one of the devices (as for all other examples in this report it is this field-dependent frequency which enables particle detection). We do note that for this device (500 nm in width), the linewidths at high fields were typically on the order of 5-20 MHz (dependent on the driving current and field).

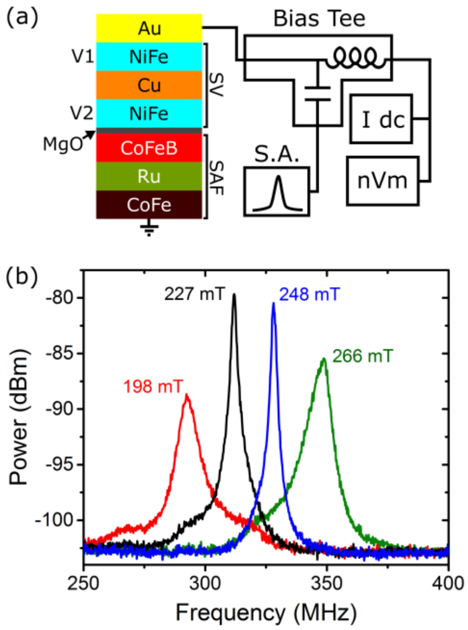

Fig. 7. (a) STO composition (vortices exist in layers V1 and V2) together with the measurement circuit (nVm=nanovoltmeter, S.A.=spectrum analyzer, I dc = low noise d.c. current source). (b) Field dependent STO output signals obtained under an 18 mA d.c. current.

[6] J.-V. Kim, Solid State Physics, 63, 217 (2012).
[7] R. Lebrun et al., Physical Review Applied, 2, 061001 (2014).

We were able to show that the STO output frequency can be modified by the presence of magnetic particles (Figs. 8a,b). This is shown for the same device discussed above. The presence of numerous particles above and around the STO (Fig. 8c) generates a clear upward shift in the device frequency (tested over a field range of ~ 100 mT and for three values of the driving d.c. current).

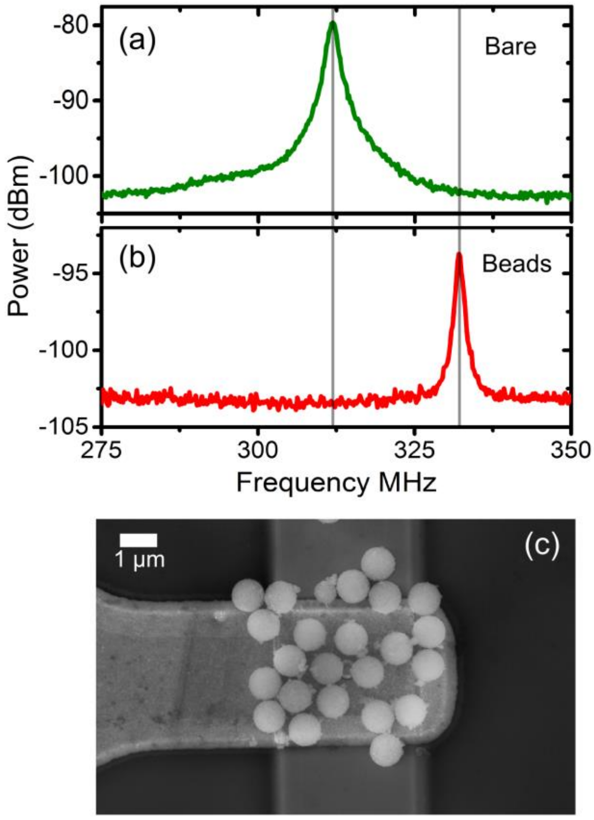

Fig. 8. (a,b) Traces showing the microbead-induced change to the STO output. (c) SEM image showing a number of beads around the device's upper contact. The STO position corresponds to the small circle which can be seen near the center of the image (between and beneath the central beads).

Note that in this device there was a significant reduction in the output power induced by the presence of the particles however this was not observed in all measured STOs. Indeed, variabilities such as this are one of the factors driving continuing work on this part of the project. We note that we have also demonstrated particle sensing using the spin diode effect (e.g. Jenkins et al.[8]). Despite a number of demonstrations of frequency-based sensing with STOs, interpretation of the signs and magnitudes of the frequency shifts remains challenging. This is potentially a result of the more complex STO structure (compared to the single disks in section 2), the presence of two (interacting) vortices in each STO and the aforementioned dependence of particle-generated gyrotropic frequency shifts on vortex chirality and imperfect field angles (section 2d). For this reason, we intend to also experimentally measure STOs in the future which are uniformly magnetized and which may yield results which are simpler to interpret. We finally

[8] A.S. Jenkins et al., Nature Nanotechnology, 11, 360 (2016).

note that a low noise amplifier has also been integrated into the setup to facilitate real time measurements (ongoing) of device outputs using an oscilloscope.

Related to the above discussion, we note that we have already shown in [2] via magnetic eigenmode simulations that STOs in which the oscillations are occurring in a magnetically uniform, out-of-plane magnetized layer (i.e. not a vortex-containing layer) have the potential to be suitable for use in nanoparticle detection. Indeed, we predict a frequency shift of the fundamental resonance mode of ~ 20 MHz at a vertical separation of 80 nm for a 20 nm wide, high moment particle. However, detection of such a shift would be reliant on achieving sufficiently low signal linewidths (discussed in more detail in [2]). In [2] we also discuss how resonant frequency shifts depend on the particle's properties, its location and the excited mode. Finally, we note that a paper using the aforementioned eigenmode method to study localized magnetic resonances in magnetic domain walls was also completed and published during the performance period [6].

Future work

Some parts of the project have been noted above to be ongoing. In terms of true *follow-on work* however, experimental studies of STOs likely represent the most critical focus to move this study forward (albeit with some simulation to aid interpretation of results). Such work would require continued collaboration with the Unité Mixte de Physique CNRS/Thales and AIST in Japan. Fortunately, the PI has, in conjunction with both groups, received a small collaboration grant (a UWA "Research Collaboration Award") to support some continued collaboration in this area in the short term (2017). In the long term however, the following activities would be most critical (in approximate chronological order):

1. Studies of experimental particle-generated frequency shifts for a very high number of STOs and for different STO types (e.g. vortex-based *and* uniformly magnetized devices) to *conclusively* determine the effect of particles on the STO output frequency and other STO signal characteristics (e.g. power and linewidth).
2. Following on from 1, we would then test STO readout methods (e.g. spin diode or real time signal readout) in terms of determining the most practical methods for rapid and/or high sensitivity detection of nanoparticles and microbeads.
3. The final goal would be focused on applying the above knowledge to enable detection of flowing particles or biological analyte detection. This would require modifications to STO contact designs, surface modifications and integration with reaction wells or microfluidics.

Note that the PI has also started building links with local medical researchers (including the submission of common grant applications) which will facilitate (in the future) in efficiently moving this work towards relevant biomedical proof-of-concept testing.

List of Publications and Significant Collaborations that resulted from your AOARD supported project:

a) papers published in peer-reviewed journals.

[1] J.P. Fried, H. Fangohr, M. Kostylev, P.J. Metaxas, "Exchange-mediated, non-linear, out-of-plane magnetic field dependence of the ferromagnetic vortex gyrotropic mode frequency driven by core deformation", Physical Review B, 94, 224407 (2016).

[2] M. Albert, M. Beg, D. Chernyshenko, M.-A. Bisotti, R.L. Carey, H. Fangohr, P.J. Metaxas, "Frequency-based nanoparticle sensing over large field ranges using the ferromagnetic resonances of a magnetic nanodisc", Nanotechnology, 27, 455502 (2016).

[3] M. Sushruth, J. Ding, J. Duczynski, R.C. Woodward, R.A. Begley, H. Fangohr, R.O. Fuller, A.O. Adeyeye, M. Kostylev, P.J. Metaxas, "Resonance-based Detection of Magnetic Nanoparticles and Microbeads Using Nanopatterned Ferromagnets", Physical Review Applied, 6, 044005 (2016).

[4] M. Sushruth, J.P. Fried, A. Anane, S. Xavier, C. Deranlot, M. Kostylev, V. Cros, P.J. Metaxas, "Electrical measurement of magnetic-field-impeded polarity switching of a ferromagnetic vortex core", Physical Review B Rapid Communications, 94, 100402(R) (2016).

[5] J.P. Fried and P.J. Metaxas, "Localized magnetic fields enhance the field-sensitivity of the gyrotropic resonance frequency of a magnetic vortex", Physical Review B, 93, 064422 (2016).

[6] P.J. Metaxas, M. Albert, S. Lequeux, V. Cros, J. Grollier, P. Bortolotti, A. Anane and H. Fangohr, "Resonant translational, breathing and twisting modes of pinned transverse magnetic domain walls", Physical Review B, 93, 054414 (2016).

b) papers published in non-peer-reviewed journals or in conference proceedings,

n/a

c) conference presentations (as presenting author),

‘Dynamic detection of magnetic nanoparticles’, MOSAIC Workshop (closing workshop of a EU FP7 project on “MicrOwave Spintronics as an AlternatIve Path to Components and Systems for Telecommunications, Storage and Security Applications”), Varberg, Sweden (June 2016) [**invited oral presentation**].

‘Towards frequency-based spintronic detection of magnetic nanoparticles’, 11th International Conference on the Scientific and Clinical Applications of Magnetic Carriers, Vancouver, Canada (May/June 2016) [oral presentation].

‘Magnonic crystals as dynamic nanoparticle detectors’, 13th MMM-INTERMAG Joint Conference, San Diego, USA (January 2016) [oral presentation].

‘Confinement-enhanced sensitivity of the vortex gyrotropic mode to localized magnetic fields’, 13th MMM-INTERMAG Joint Conference, San Diego, USA (January 2016) [poster presentation].

‘Vortex core polarity switching evidenced via signal loss during magnetoresistive spectroscopy’, 13th MMM-INTERMAG Joint Conference, San Diego, USA (January 2016) [oral presentation].

‘Frequency-based magneto-electronic nanoparticle detectors: Towards fast, nano-scale, spintronic biosensors’, Theo Murphy Australian Frontiers of Science symposium (‘Materials for the 21st century: from design to application’), Melbourne, Australia (December 2015) **[invited oral presentation]**.

d) manuscripts submitted but not yet published, and

[7] J.P. Fried and P.J. Metaxas, "Nanoparticle-modified magnetic vortex dynamics", accepted for publication in IEEE Magnetics Letters (2017).

[8] M. Sushruth, J.P. Fried, A. Anane, S. Xavier, C. Deranlot, V. Cros and P.J. Metaxas, "Chirality-mediated bistability and strong frequency downshifting of the gyrotropic resonance of a magnetic vortex", under review at Physical Review Letters (2017).

e) provide a list any interactions with industry or with Air Force Research Laboratory scientists or significant collaborations that resulted from this work.

This work has involved collaborations with scientists and engineers at Thales Research and Technology (Palaiseau, France) and the Unité Mixte de Physique CNRS/Thales (also located within the Thales Research and Technology facility in Palaiseau).

We also received nanoparticles from Senior Scientific as part of their TrailBlazer Trial Program.